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Title: Spray Drying of High Explosives

Author(s): Tisdale, Jeremy Tyler
Hill, Larry Glenn
Duque, Amanda Lynn

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Los Alamos NATIONAL LABORATORY

Spray Drying of High Explosives

Jeremy Tisdale, Larry Hill, Amanda Duque
Q-5: HE Science & Technology

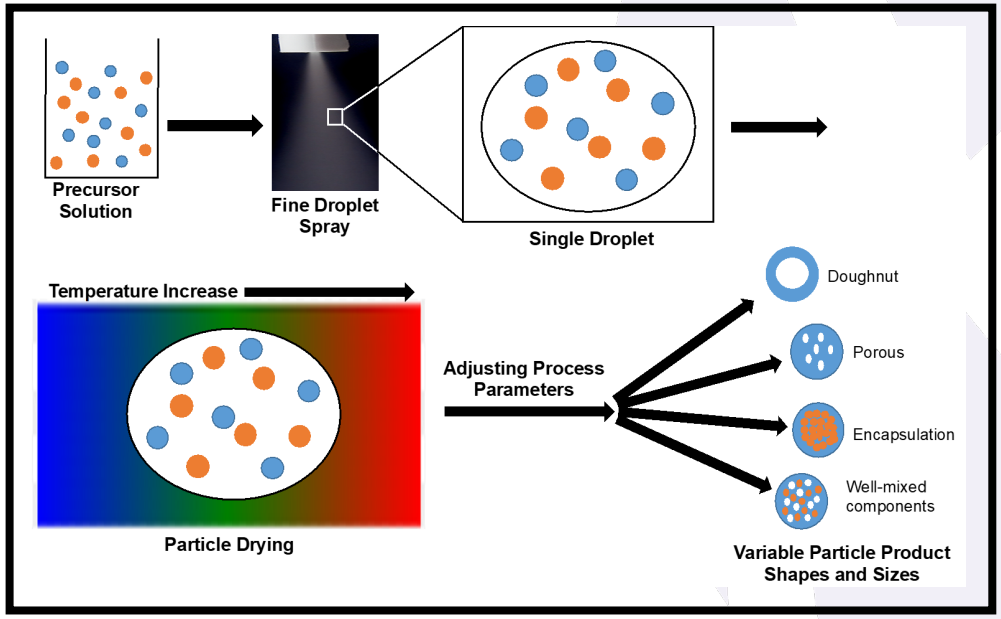
08/25/21

Presentation Outline

1. What is spray drying?
2. How can spray drying be utilized for high explosives processing?
3. Literature overview.
4. Examples of current spray drying research done in Q-5.
5. Future plans and outlook for spray drying high explosives.

Spray drying offers versatile options for HE production and formulation.

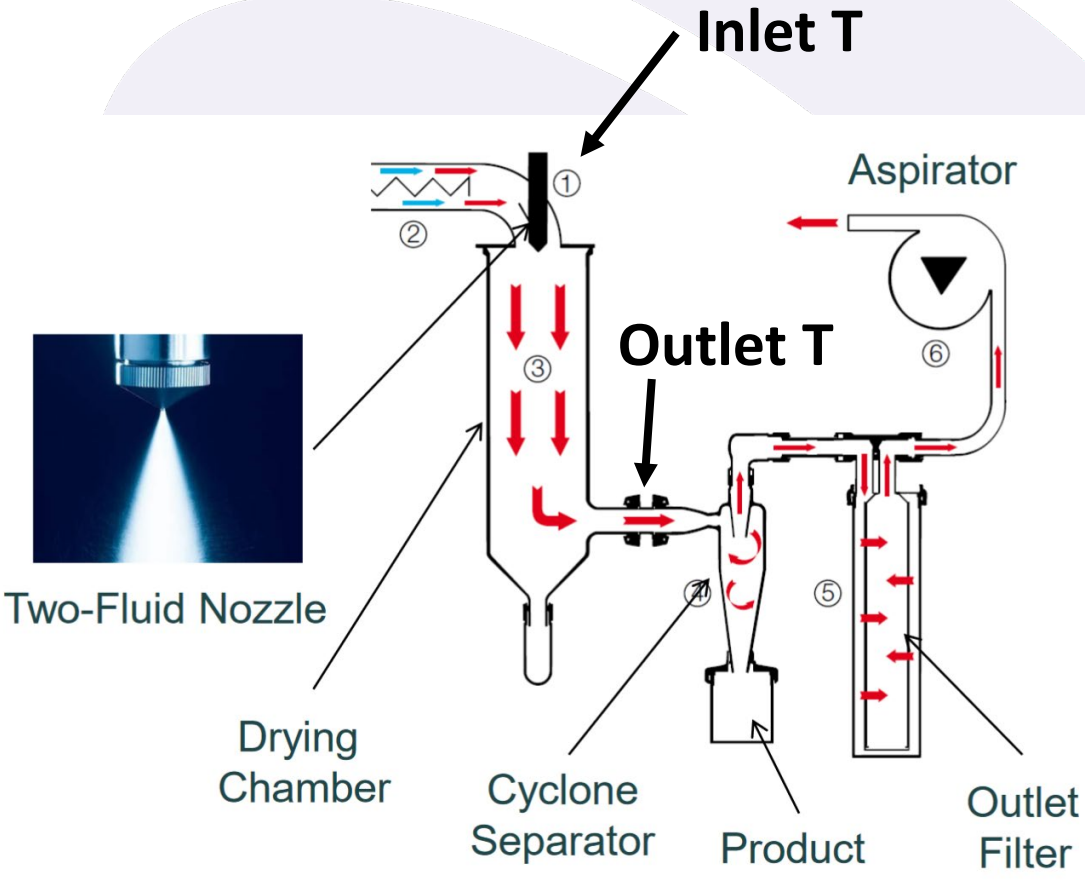
- Spray drying is an industrial technique where a solution is sprayed through a hot drying medium, resulting in dry, fine powders.
- Benefits for HE include:
  - Simple, one-step process for producing uniform nano/micro-HE particles and formulations
  - Control/tuning of particle size and morphology (sub-micron, up to ~50 μm)
  - *Allows for access to HE microstructures that we do not currently have by any other production means*



Benchtop laboratory unit with opportunities for scale-up

On-going objectives focus on understanding the parameter space for control over the final product size and morphology.

- Range of neat HEs and formulations
- Temperature gradient, gas and feed flow rates, yields of material, feedstock concentration
- 4 different spray nozzles:
  - 0.7, 1.4, and 2.0 mm opening atomization nozzles
  - ultrasonic nozzle
- Solvent choice → dictates needed outlet temperature
  - Outlet temperature based on boiling point of solvent for full drying of material



How can spray drying be utilized in high explosives processing?

Spray Drying

Feed

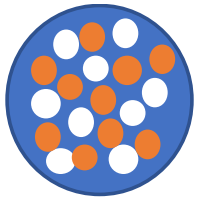
HE + binder dissolved in solution

Product

Neat HE with unique particle size/morphology

Well-mixed composites or co-crystals

Particle size of product may be tuned by processing parameters



Spray Coating

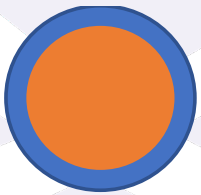
Feed

Slurry of undissolved HE + dissolved binder

Product

Micro-encapsulated HE with binder

Particle size/morphology of the starting material is retained in the product (FEM-HMX, UF TATB, etc.)



Reported Literature Spray Drying Efforts

- Considering the small amount of groups (only a few world-wide) using this capability, a significant amount of research has been reported on a variety of spray dried HE materials, composites and formulations
- One-step spray drying method for HE/Binder composites
  - HMX/Estane, HMX/Viton, HMX/PVAc, HMX/PVOH, RDX/PVAc, RDX/Estane CL-20/EPDM
- Multi-HE composites and co-crystallization
  - HMX/Nitrocellulose, HMX/TATB, HMX/TNT, CL-20/HMX, CL-20/DNDAP
  - Lack of single crystal crystallography data, questions still remain for proven successful co-crystallization
- Novel amorphous energetics
  - CL-20/HMX/PVAc

Qiu, H.; *et al., Powder Tech.*, 274(2015) 333-337.

Ye, B.; *et al., RSC Adv.*, 7(2017) 35411-416.

Liu, N.; *et al., CrystEngComm.*, 20(2018) 2060-67.

Stepanov, V.; *et al., Propellants, Explos. Pyrotech.*, 41(2015) 142-147.

Current Goals for Spray Drying HE

- One-step processing of PBX nanocomposites
- Fully dissolved solutions of HE/Binder can be spray dried to achieve size and morphology controlled HE composites
  - HMX/Estane, RDX/PVAc, etc.
- One-step co-crystallization for useful HE composites
  - For example, spray drying CL-20/HMX co-crystals showed a drastically reduced sensitivity as compared to pure CL-20 or pure HMX. An, C., *et al.*, *Journal of Nanomaterials* 2017(2017).
- Spray drying of neat HE materials for size reduction and morphology control

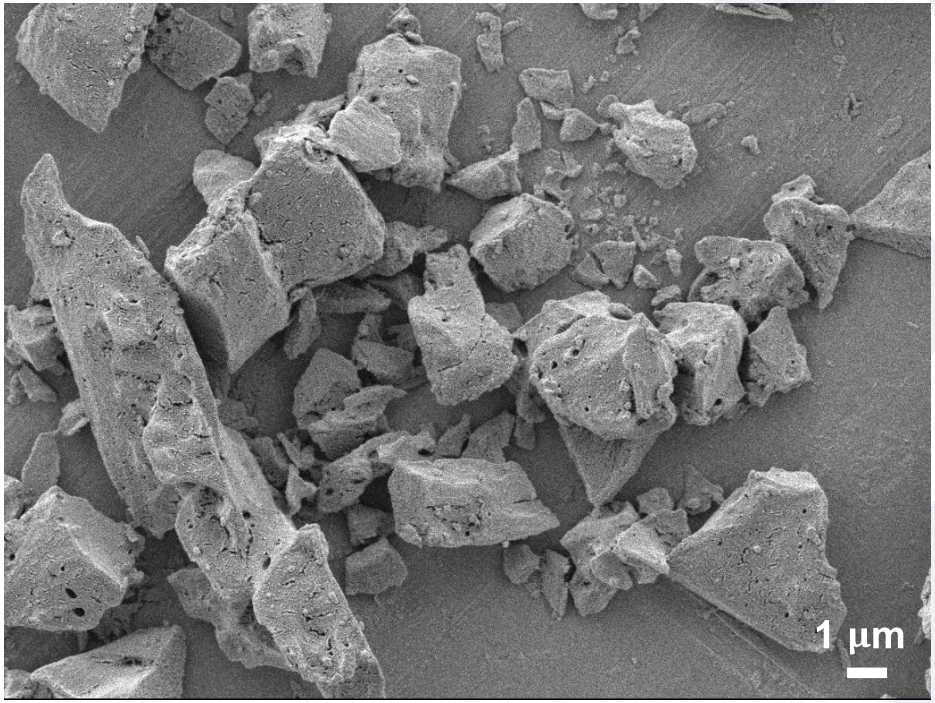
Current Materials Undergoing Research Efforts

- Neat PETN (LA-UR-21-23797)
- Neat TNT
- Neat RDX
- HMX and PBX-9501
- PFBA (formulated as PBX-9501) and other mocks to understand processing conditions

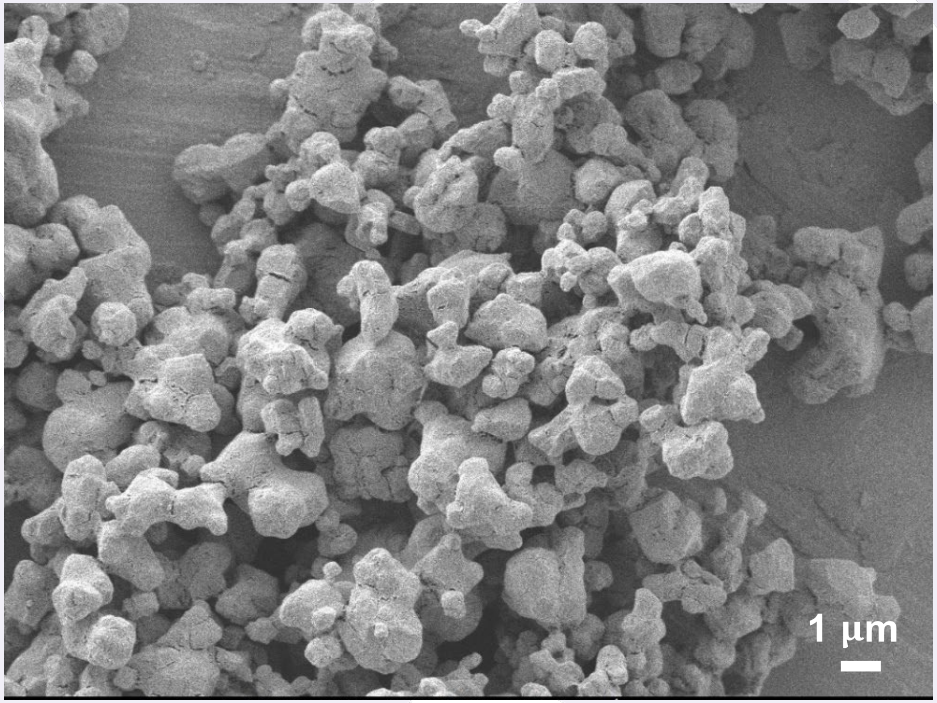
Spray Drying PETN

- Spray drying PETN has provided a novel size/morphology for this material
- Size reduction is a known method to decrease sensitivity
- Spray drying used as a non-mechanical processing technique for sensitive explosives

Starting Material: PETN XTX Grade

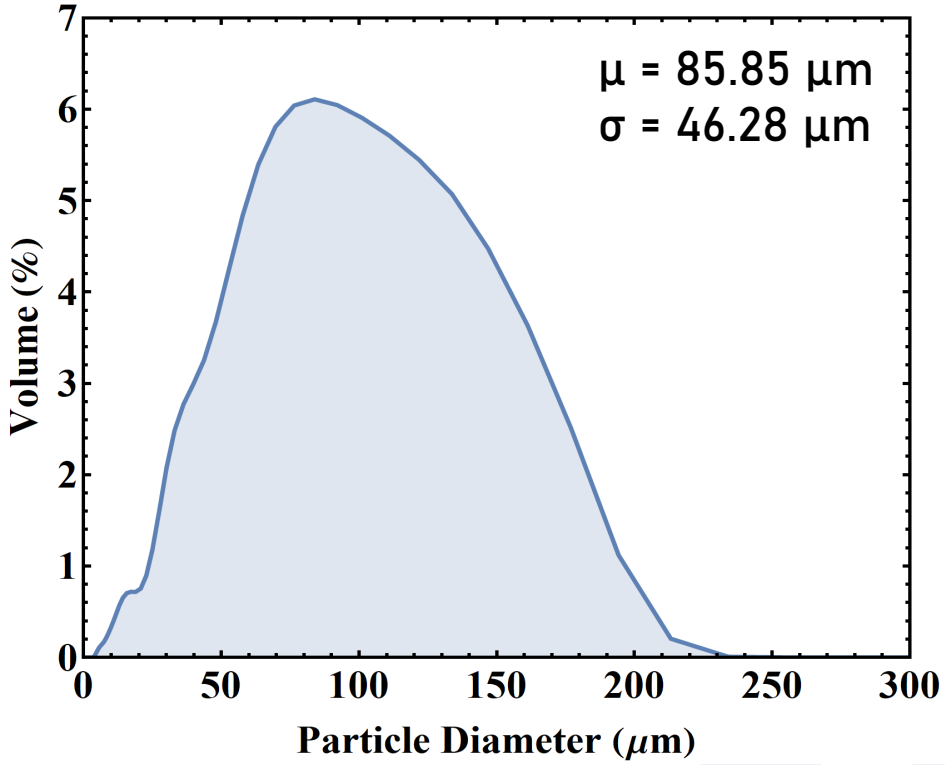


Spray Dried: PETN (Neat)

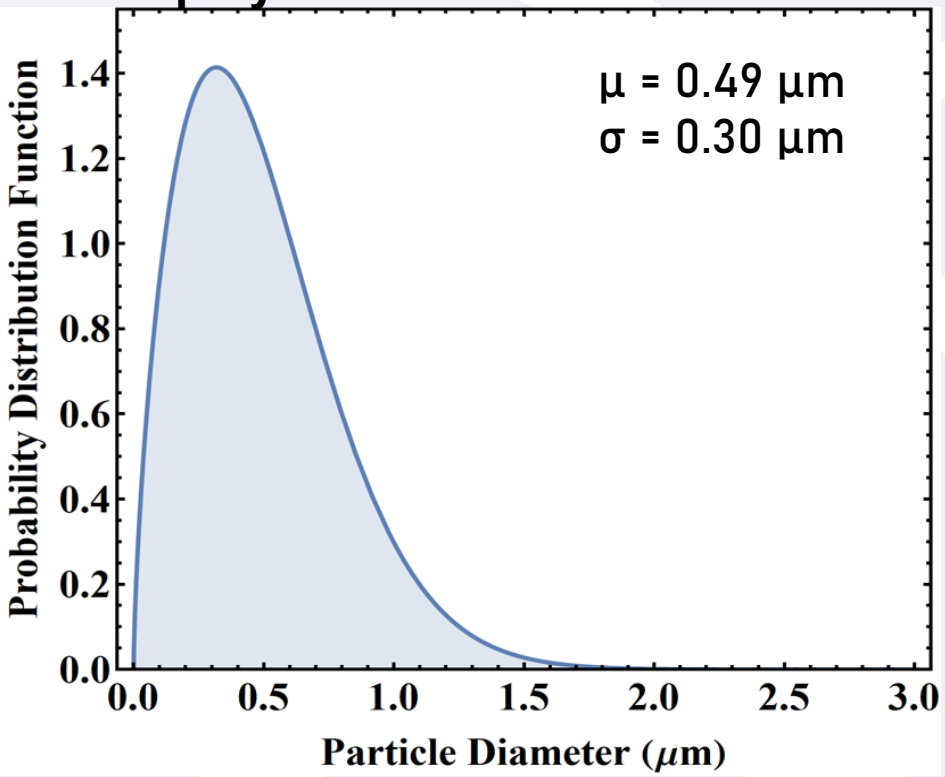


Physical Characterization of Spray Dried PETN

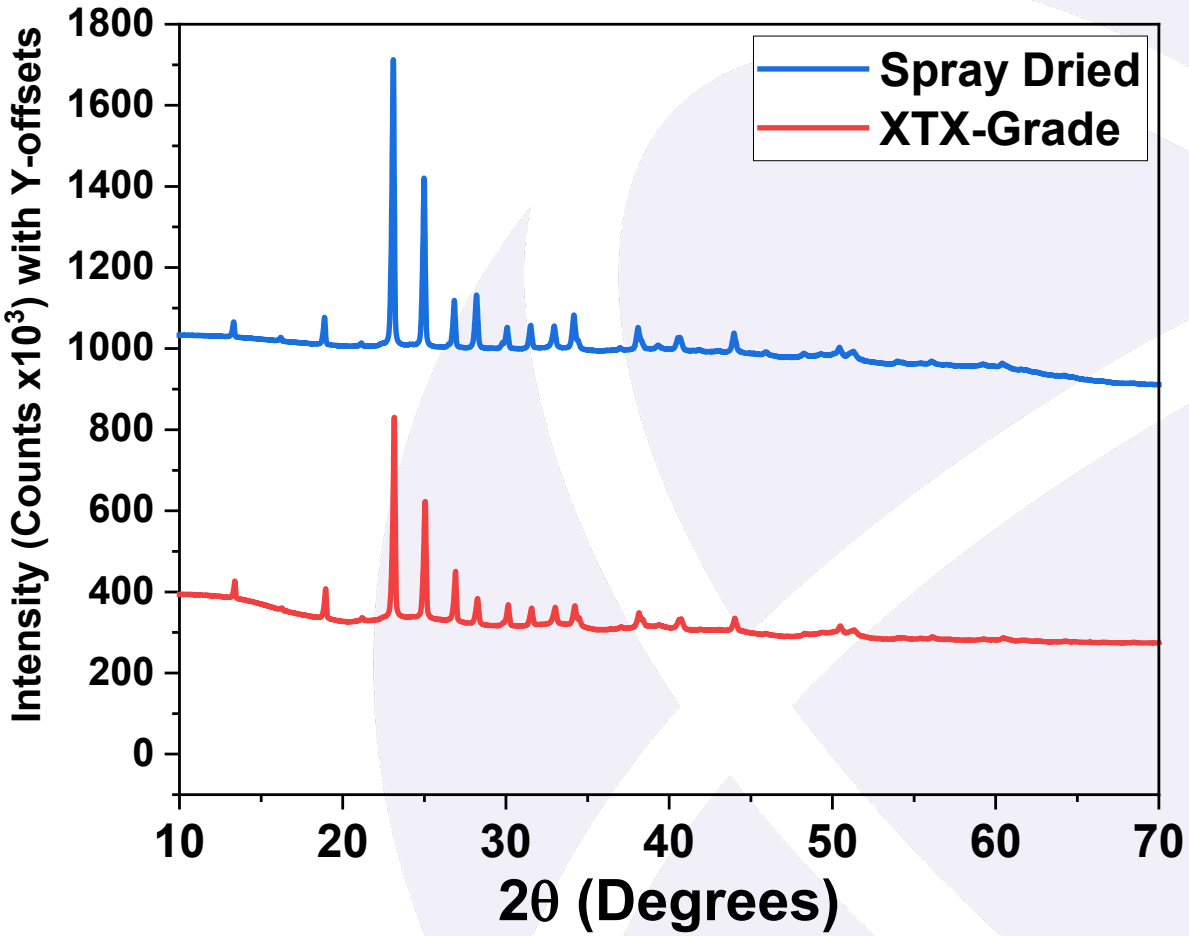
Coulter PSD Analysis
XTX-Grade Standard PETN



Keyence Image PSD Analysis
Spray Dried PETN



Physical Characterization of Spray Dried PETN



- Powder XRD shows that the powder PETN remains in the thermally stable, tetragonal, PETN-I polymorph

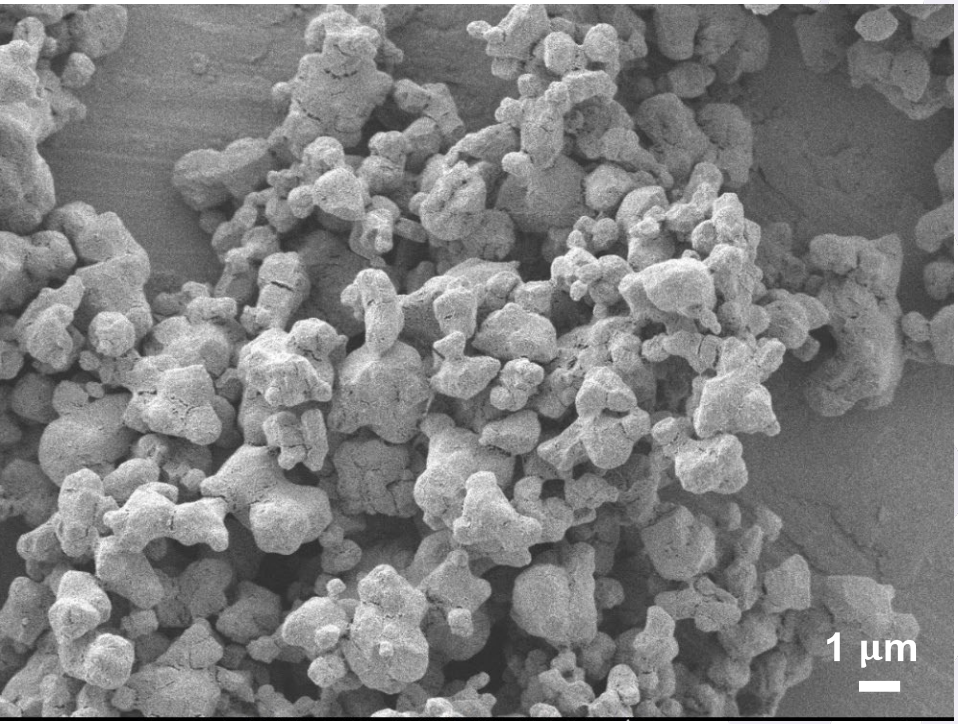
Small-scale sensitivity of PETN

Sample	Impact Testing		Friction Testing		ESD Testing	DSC Results	
	50% Height (cm)	σ (cm)	50% Load (N)	σ (N)	*TIL/**Screen (J)	Melt T (°C)	Onset T (°C)
PETN XTX Grade	11.1	1.3	134.7	17.4	*0.0625	140.6	165.4
PETN L298 Standard	11.8	1.7	60.1	3.6	*0.125	140.8	165.8
PETN Spray Dried	18.1	3.0	201.5	58.8	*0.125	140.8	165.8

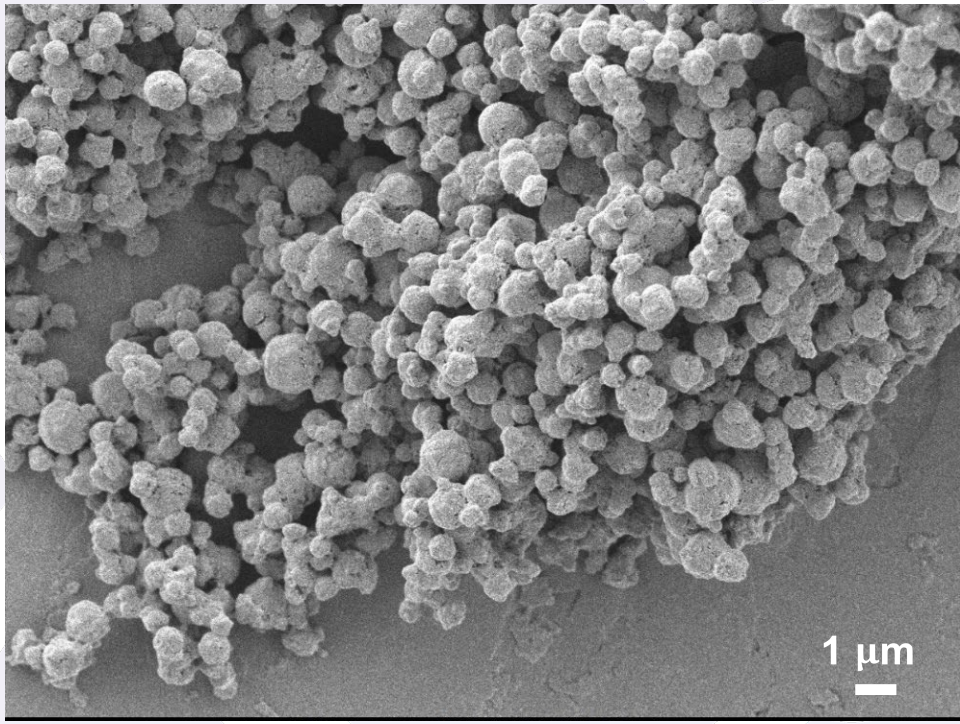
- DSC results are used to ensure no undesirable chemical properties are changing such as the onset of decomposition
- Small-scale sensitivity tests showed a reduction in impact and friction sensitivity!
- Further detailed information: LA-UR-21-23797

The presence of an additive or binder will have a significant effect on the product morphology

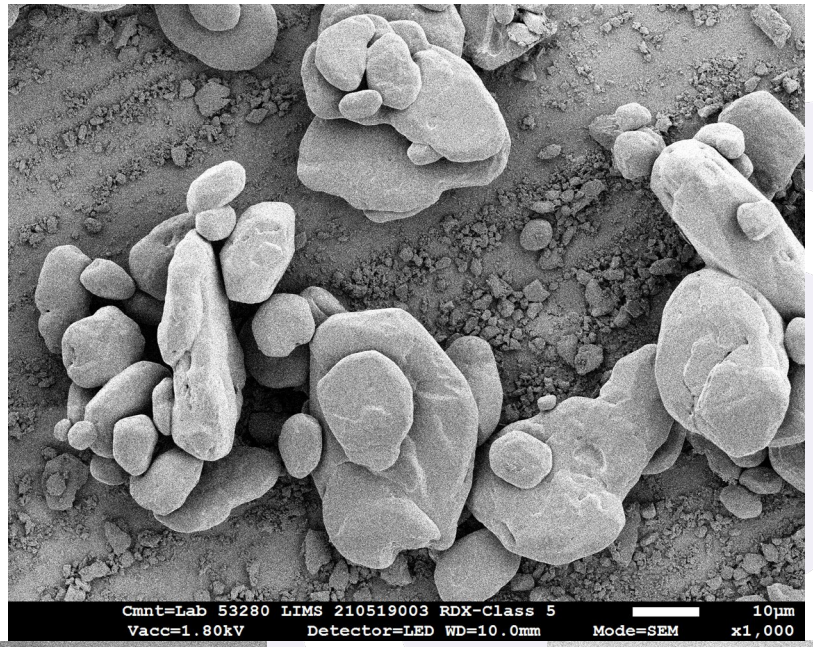
Spray Dried: PETN (Neat)



Spray Dried: PETN w/ 5 wt% Kel-F

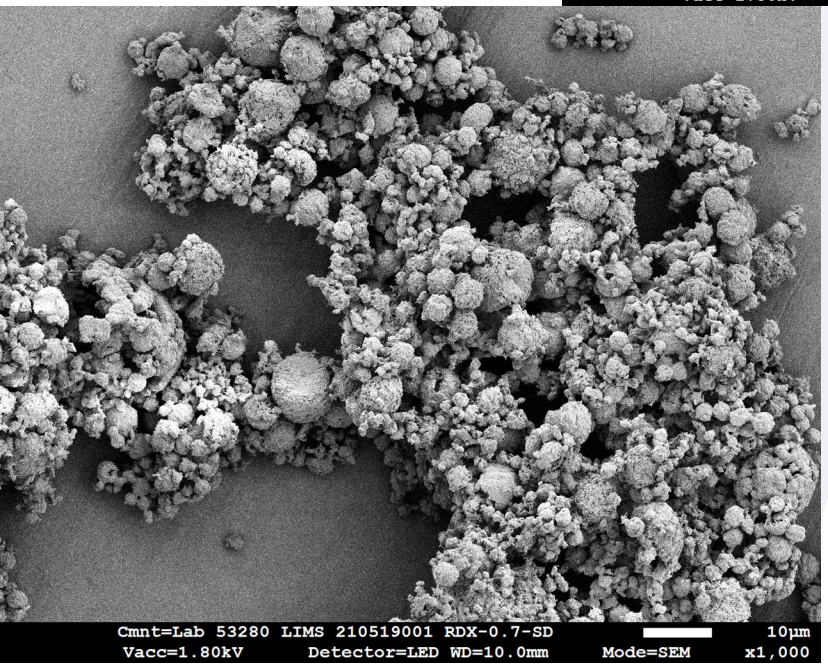


Spray Drying RDX

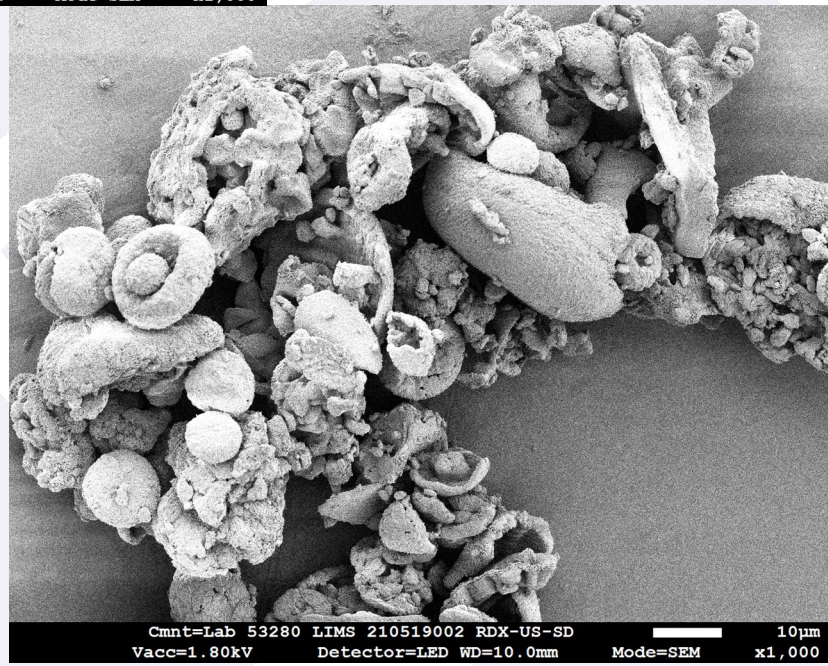


Class 5 RDX
Standard Starting
Material

Spray dried with
0.7 mm atomizing nozzle

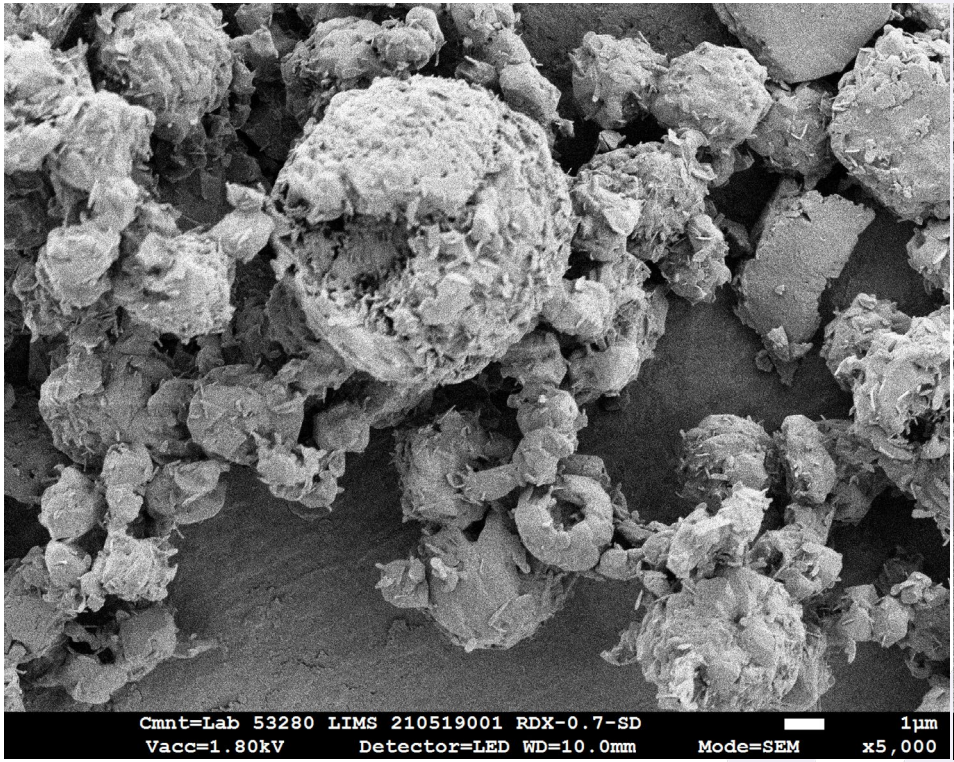


Spray dried with
Ultrasonic Nozzle

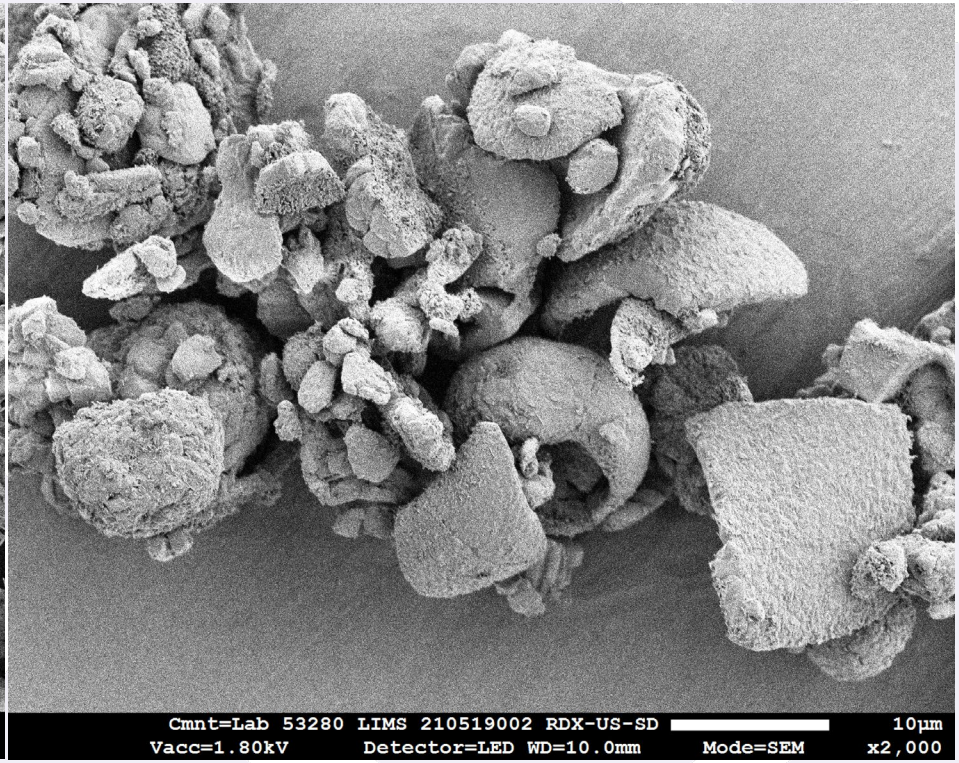


Spray Drying RDX – Spray Dried Surfaces

Spray dried with
0.7 mm atomizing nozzle



Spray dried with
Ultrasonic Nozzle



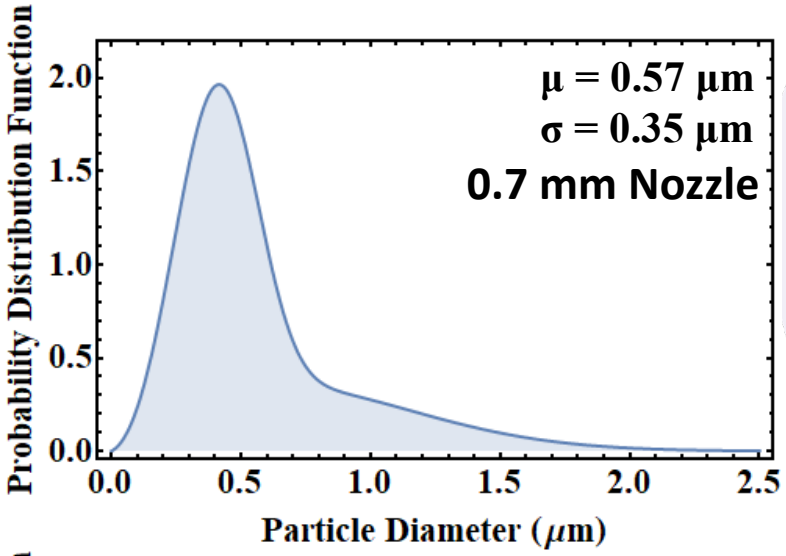
Small-scale sensitivity of RDX

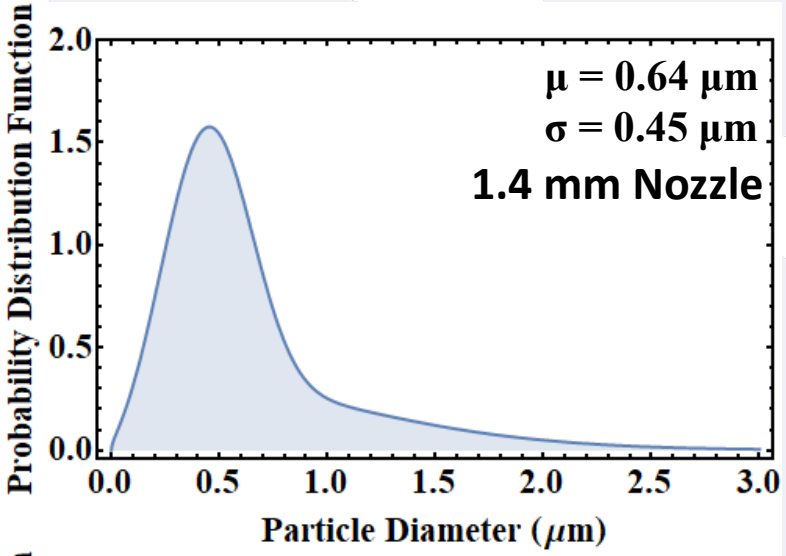
Sample	Impact Testing		Friction Testing		ESD Testing
	50% Height (cm)	σ (cm)	50% Load (N)	σ (N)	*TIL/**Screen (J)
RDX Class 5	30.2	1.0	>360	NA	*0.125
RDX – Spray Dried US	25.5	1.9	>360	NA	*0.0625
RDX – Spray Dried 0.7	22.8	2.7	296.9	78.2	*0.125

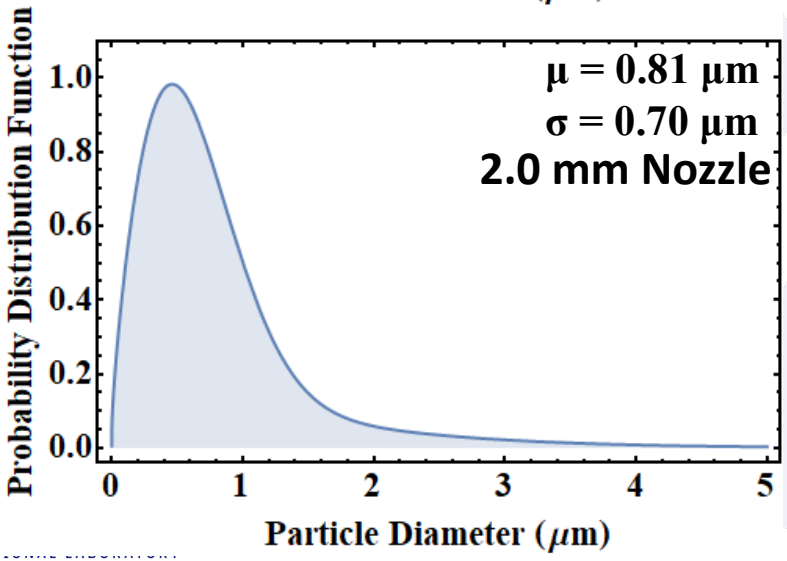
- Unlike the PETN, the spray dried RDX showed increased sensitivity to impact and friction as we decreased the PSD
- The increase in sensitivity is most likely due to the morphological changes
- Work is continuing to understand the cause of this trend

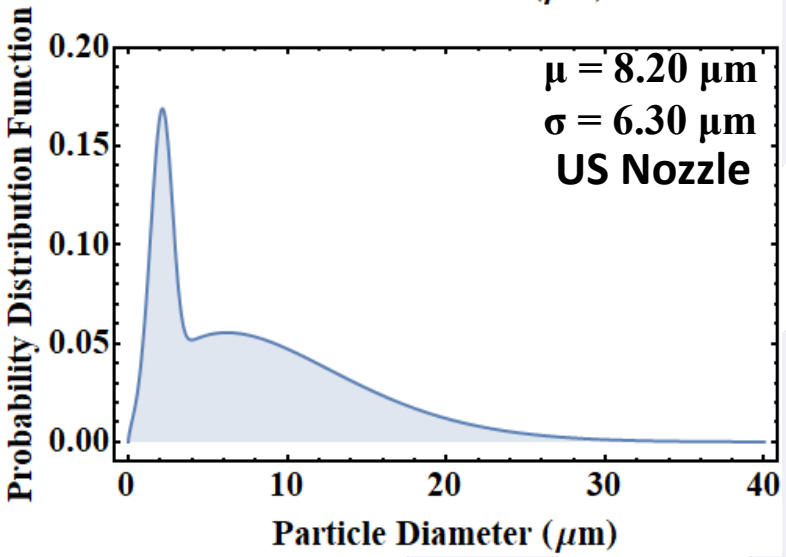
Spray Drying RDX

- Neat RDX as a test case system to understand PSD tuning with processing conditions

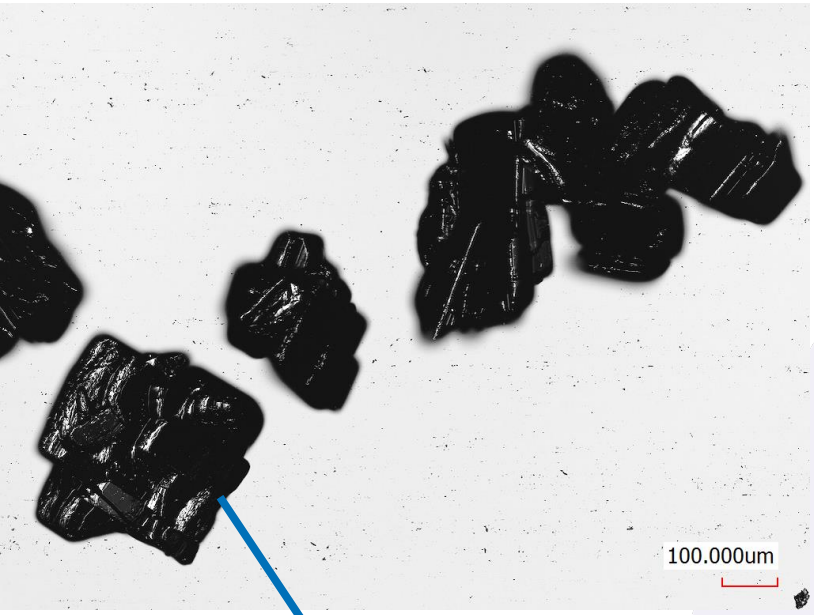


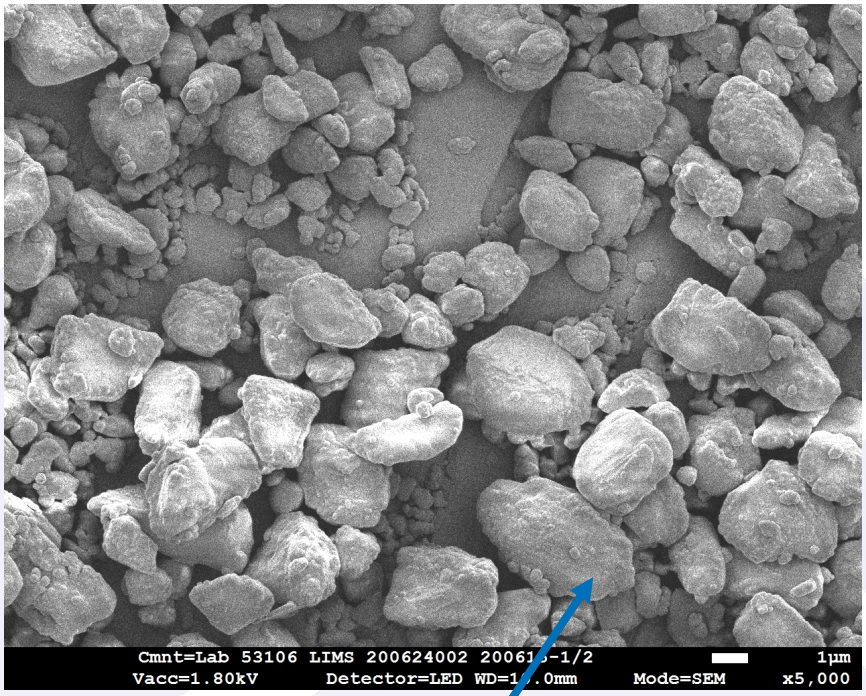






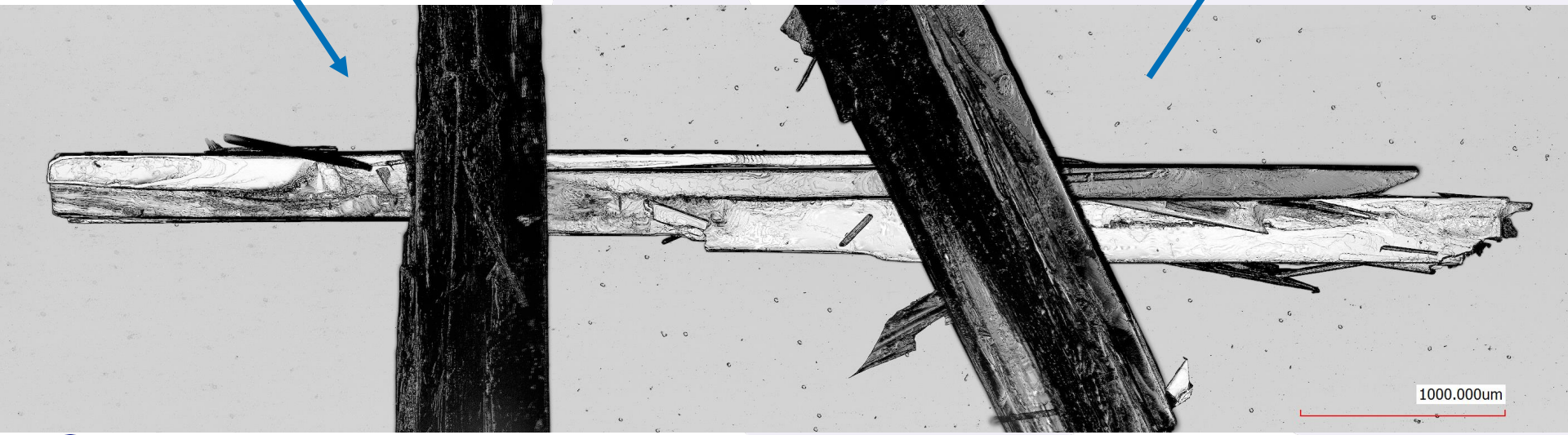
Spray Drying TNT



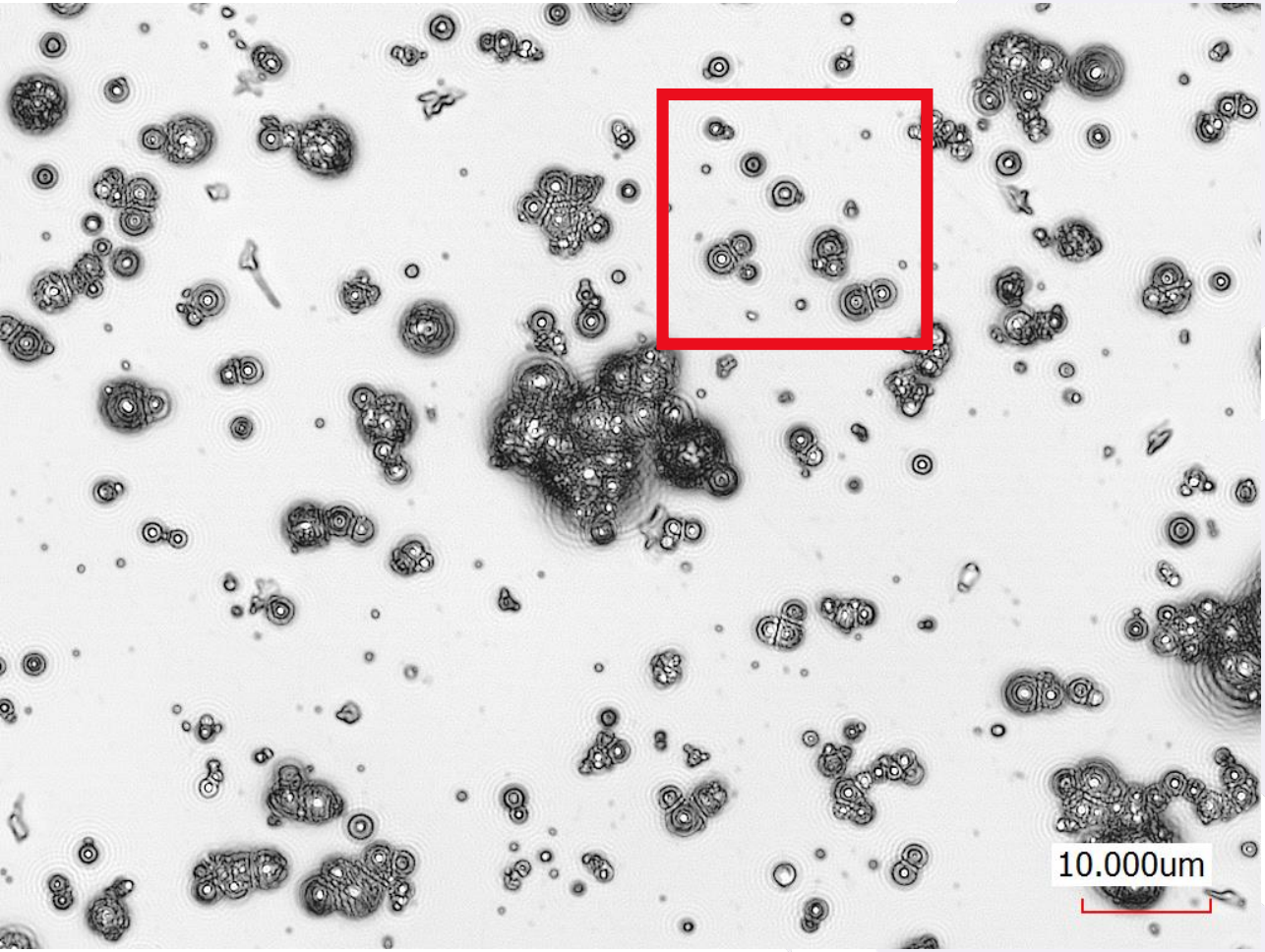


Crash-out Purification

Spray Dried



Spray Drying HMX/Estane



- Initial spray drying experiments with 95/5% HMX/Estane
- These experiments will look at microencapsulation as well as spray drying full dissolution of components
- Plan to start experimenting with added nitroplasticizer

On-going and Future Work in Spray Drying HE

1. Continue to develop understanding of processing conditions to have accurate, fine-tuned control of particle size distributions and morphologies.
2. Continue to investigate neat explosives with novel morphologies/microstructures and understand the impact and connections with physical, chemical, and energetic properties.
   - This includes PETN, HMX, RDX, TNT, etc.
3. Begin experimentation with microencapsulation of PBX systems to explore coating uniformity, and material stability, as well as alternate binders viable with spray drying
4. Begin studying processing of multi-component HE systems to explore potential co-crystallization processes and novel energetic formulations

Thank you!

List of Reports:
Characterization of Spray Dried PETN Powders (LA-UR-21-23797) (report)
Production of Desensitized, Ultrafine PETN Powder (waiting for submission)
Investigation of Neat Spray Dried RDX Powder Morphologies (In preparation)
Effects of Spray Drying on TNT Polymorphism (In preparation)